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Abstract

UPON THE PHYSICOCHEMICAL LAWS GOVERNING THE DISPOSAL OF SOME HEAVY RADIOELEMENTS INTO THE GROUND

M.A. El-Guebeily and N.Z. Misak
Atomic Energy Establishment, U.A.R.

A series of studies have been previously undertaken in UAR.AEE to examine the feasibility and the characteristics of disposal of radioactive wastes of Sr, Ru, Cs and Ce into the ground. The present work aims at complementing these studies by data upon the behaviour of some heavy alpha emitting radioelements and in particular to examine the physicochemical laws governing their interaction with sand and clay minerals. For this purpose uranium and thorium were chosen in the forms of uranyl nitrate in HNO_3, uranyl carbonate complex in Na_2CO_3 solution and thorium nitrate in HNO_3; the concentration of U and Th being of the order of 10^{-3}M. As adsorbing materials, Inshas reactor site sand and an Egyptian vermiculite were used. Both batch and column techniques were applied in the uptake experiments. The percentage decontamination and the pH value were followed with time till after equilibrium. The effects of initial pH, grain size, temperature and concentration on the uptake process were investigated. The change of concentration of U and Th was followed spectrophotometrically.

The results obtained indicate that:

(a) Uranium in the anionic form is not taken up by sand and is only slightly absorbed by vermiculite.

(b) UO_2^{2+} and Th^{4+} are very slightly taken up by largest size sand.

(c) UO_2^{2+} and Th^{4+} uptake by finest and intermediate size sand, as observed from the kinetic curves, is rapid for relatively low initial pH values, but a second slow process appears where the pH of the solution exceeds the value of 2.

(d) The uptake of Th^{4+} by intermediate and finest vermiculite sizes is very rapid, in contradistinction to UO_2^{2+} for which equilibrium is attained in about 30 minutes. With largest size, the results are irreproducible.

(e) H^+ is taken up in parallel with UO_2^{2+} and Th^{4+}. The capacity for UO_2^{2+} and Th^{4+} increases with increases of initial pH and with decrease of particle size.

(f) The effect of temperature on uptake is minor in all cases.

In order to correlate and interpret the main uptake data, the following physicochemical treatment was tried:

1 - To evaluate the number of processes involved in the uptake phenomena, the equation of unimolecular equilibrium

$$-(k_1 + k_2) = \frac{1}{t} \ln\left(1 - \frac{x}{x_e}\right)$$

was used as an approximation, neglecting other influences. This equation has the form of McKay's equation

$$k = -\frac{ab}{a+b} \cdot \frac{1}{t} \ln\left(1 - \frac{x}{x_e}\right)$$

with difference however in the physical meaning of the constants. The number of processes obtained is in accordance with the kinetic curves on sand and the known properties of vermiculite.

2 - The evaluation of the equilibrium distribution coefficient

$$k_d = \frac{\% \text{ adsorbed}}{100\% - \% \text{ adsborbed}} \cdot \frac{V}{M}$$

at different acidities is in favour of an ion exchange adsorption mechanism.

3 - To evaluate the rate determining step, the particle diffusion equation

$$Q_t/Q_e = 6Q_o\sqrt{\frac{Dt}{\pi}} \;/\; r\,(Q_o - Q_e)$$

was applied and it was found that in all cases the main uptake processes are most probably particle diffusion controlled. The variation of the internal diffusion coefficient with pH is indicative of changes in the structure of the adsorbable ionic species.

4 - The data on sand may be described quantitatively by Langmuir isotherm

$$\frac{C}{x/m} = \frac{1}{k_1 k_2} + \frac{C}{k_2}$$

while those on vermiculite may be correlated by a Freundlich isotherm

$$\frac{x}{m} = kC^{\frac{1}{n}}$$

with more than one deflection, indicating the possible presence of different energy sites.